INDEX OF REFRACTION OF NICKEL AT HIGH TEMPERATURES

by

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INTRODUCTION

Early in the history of x-radiation the electromagnetic nature of x-rays was determined. In the years following it was discovered that the absolute index of refraction associated with x-rays was less than unity. Because of the proximity of the value of this index to one, a quantity δ is usually tabulated where

$$\delta = 1 - \mu$$

and μ = index of refraction.

Compton (4) was the first to make use of the fact that μ is less than one to show that x-rays would undergo total external reflection from a polished surface in a manner similar to the total internal reflection that was well known in the longer wavelength region of the electromagnetic spectrum.

Total external reflection was used to determine the value of δ by applying Snell's Law. At the critical angle, the angle of refraction is 90 degrees and Snell's Law becomes

$$\sin i_c = \mu$$

where i is the angle of incidence. In terms of the glancing angle θ, this may be written

$$\cos \theta_c = \mu .$$

Expanding the $\cos \theta_c$ in a power series and neglecting higher powers of θ_c there results

$$\tfrac{1}{2} \theta_c^2 = 1 - \mu = \delta .$$

Thus by measuring θ_c, investigators have been able to determine δ directly.

With the discovery of the electron and the development of the electron theory, Lorentz (12) determined a theoretical expression for the index of refraction yielding a complex value for μ. The real part of the complex index gives the ratio of the velocity of the wave in a vacuum to the velocity of the wave in the material and is called the refraction index. The real coefficient of the imaginary part of the complex index gives a constant times the linear absorption coefficient and is called the absorption index. The derivation of these equations may be found in any standard theoretical or x-ray text such as Compton and Allison (4). The final result is tabulated in the following equations:

$$\mu = 1 - \delta - i\beta = 1 + \frac{2\pi e^2}{m} \sum_q \frac{n_q}{\omega_q^2 - \omega^2 - ik\omega^3}$$

where $k = 2e^2/3mc^3$

c = velocity of light in cm per second

e = charge on electron in esu

m = mass of the electron in grams

ω = circular frequency of the incident radiation

ω_q = natural circular frequency of q type electrons

n_q = number of type q electrons per cubic cm

Although the term total reflection has been applied to this phenomena, the reflected intensity has been found to be a function of the angle of reflection and of the absorption. From the Fresnel equations for reflection it has been shown that the intensity of reflection at a particular angle depends on the ratio of β to δ (8). Kiessig (10) found experimentally that the sharpest cutoff at the critical angle occurred when the frequency of the incident radiation was just less than the frequency

of the K absorption edge of the material being investigated. The values of δ and β obtained from the Lorentz theory of dispersion did not predict this nor has the Lorentz theory given values of δ that compared with experimental data in the region of the critical absorption frequencies.

By assigning a set of virtual oscillators to each group of electrons with characteristic frequency ω_q, Kramers, Kallmann and Mark obtained expressions for δ and β that compared more closely with experiment. As has often occurred, a quantum mechanical derivation by Hönl has given a theoretical result that best fits the experimental data at the present time. The theories of Kramers, et al and of Hönl involve too much detail to be reproduced here but may be found in James (8) or Compton and Allison (4).

In 1949 Lee (11) initiated an experimental investigation of the variation of the index of refraction as a function of temperature. Using a film technique, Lee was able to obtain a plot of the coefficient of expansion for nickel as a function of temperature because of the dependance of δ on the density of the material. It was desired to improve on Lee's apparatus with the aim of determining reflected intensities throughout the region of total reflection. Thereby, both δ and β could be obtained by plotting the intensity against angle of reflection and using the Fresnel equations.

This work was undertaken to make a determination of the free electron density as a function of temperature. St. John (13) and Cardwell (2) indicated from Fowler plots (5) of photoelectric data that there was

a change in the free electron density in the vicinity of the Curie temperature of iron and nickel. In the case of nickel, it was found that the number density of free electrons decreased beyond the Curie point while it remained nearly constant up to that temperature. Hence, it should be possible to analyze this data in the light of the index of refraction information since changes in δ and β would arise from a change in the overall density of nickel and, possibly, from a change of electrons from a free state to a bound state.

EXPERIMENTAL APPARATUS AND PROCEDURE

The apparatus consisted essentially of a double crystal monochromator set in the antiparallel position for Co $K_{\alpha 1}$ radiation, an evacuated furnace in which the nickel mirror was mounted and oscillated, and a side window Geiger tube and recording unit to determine the intensity of the reflected radiation. The reflected intensity was first obtained as a function of the time. The intensity was then plotted as a function of the glancing angle by means of a curve relating glancing angle and time. The complex index of refraction was then determined from the curve of glancing angle versus time, the equation relating δ and θ_c, and the Fresnel equations.

The Furnace and Sample Holder

The details of the construction, dimensions and operation of the

furnace and sample holder are to be found in the unpublished thesis of Lee (11). A brief description of this apparatus and an indication of the changes in design are here noted.

The nickel mirror was clamped in position by means of one fixed and one adjustable facing plate. The facing plates were attached to a table which was oscillated through small angles by a shaft that was rigidly connected to the table. The face of the fixed plate was situated directly above the center of the shaft and a small hole was drilled in the center of the table in which an aligning needle could be placed.

The furnace was constructed with the bottom open so that the sample could easily be removed by lifting the furnace from the table. Nichrome wires were placed in brass heating plates on the sides of the furnace and the exterior was covered with asbestos in an effort to obtain the most uniform temperature possible in the interior of the furnace. The ends of the furnace contained small openings for the transmission of the incident and reflected radiation.

The table and furnace were enclosed in a chamber of brass that was made vacuum tight by means of a cover plate in which a double shoulder was cut. A Wilson seal was employed where the drive shaft passed through the chamber. So that expansion would not cause vacuum leaks, copper tubing was soldered to the chamber and cover plate for the purpose of carrying cooling water. The chamber was equipped with two one-thousandth inch aluminum windows large enough to permit the transmission of the incident and reflected radiation.

The table was rotated by means of an arm affixed to the shaft. The

arm was made to ride against a cardioid cam that was powered by a 40 rpm motor and a series of two gear reduction boxes. The resulting rate at which the cardioid cam turned was 0.00763 rpm or one revolution every 131 minutes.

It was desired that the rotation of the mirror be a linear or nearly linear function of time in order that the curve of intensity of the reflected radiation versus the glancing angle could be easily and accurately obtained. The cardioid cam was designed to fulfill this requirement. However, the need for high precision machining was alleviated by plotting a calibration curve. The data for the calibration of angle of rotation versus time were obtained by placing the nickel mirror in position, reflecting a collimated beam of light from the mirror, and at suitable intervals of time noting the deflection on a glass scale placed across the room. As the angles of rotation were small, the tangent of the angle was replaced by the angle itself. The resulting calibration is shown in Plate I.

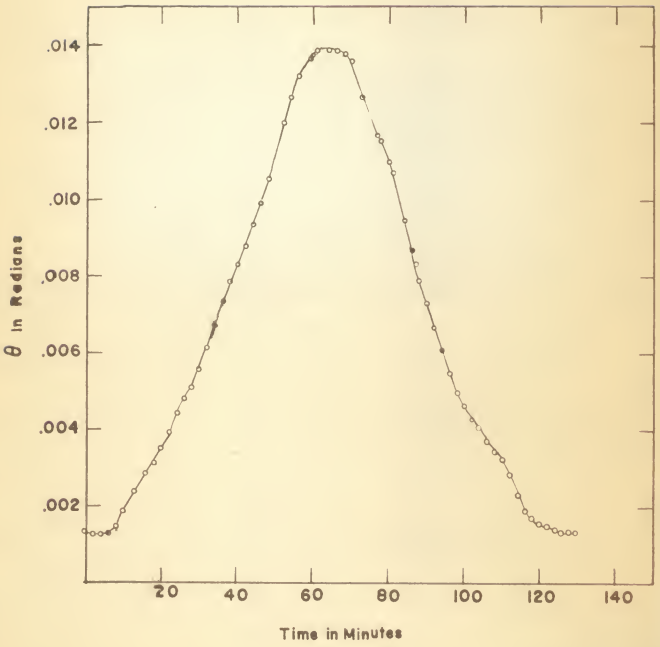
The Side Window Geiger Tube

When using non-film techniques to measure the intensity of a collimated beam of x-radiation that has been reflected, diffracted or scattered through some angle, it is usually necessary to employ a mechanism to locate the detector in a specific position with respect to the deviated beam. Ordinarily a slit is placed in front of an end-window counter and rigidly fixed to it to assure that the same region of sensitivity of

EXPLANATION OF PLATE I

The oscillation of the nickel mirror in radians is plotted as the ordinate. The time of rotation of the cardioid cam is plotted as the abscissa.

PLATE I


.014
.012
.010
.008
.006
.004
.002
θ In Radians
20
40
60
80
100
120
140
Time in Minutes

the counter is used in each measurement. This slit and counter are then moved together to analyze the angular distribution of x-ray intensities. The side-window Geiger tube was constructed to eliminate the use of such a mechanism for beams deflected through angles up to 20 degrees.

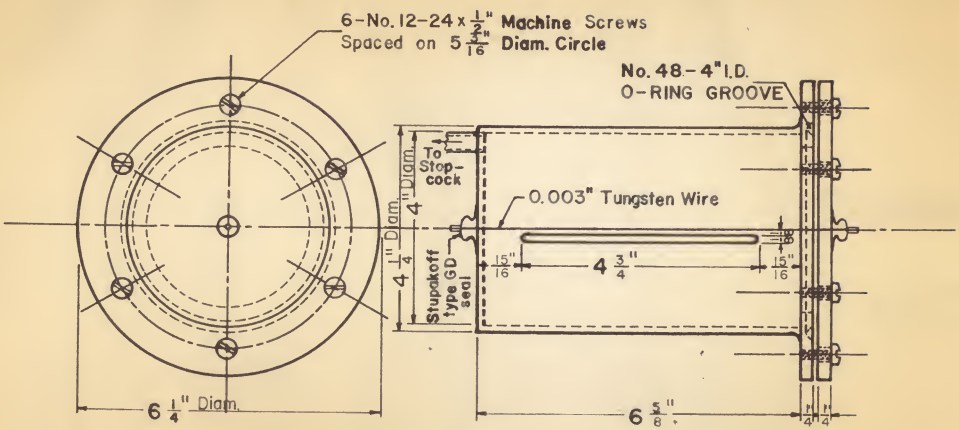
For a given x-ray beam to be recorded as the same intensity, even though the beam enters a counter at discrete but different regions of the counter window, it is required that the beam travel similar path lengths through similar electric field intensities within the counter. The latter factor becomes the more important for low energy gamma rays and x-rays. To obtain a uniformity of field intensity for each path, the Geiger tube was made 18 cm long and 10 cm in inside diameter. It was constructed of 1/8th inch brass tubing, with a limiting window 3 mm wide and 12 cm long cut along the tube parallel to the central wire. The window was covered with one thousandth inch aluminum and situated so that horizontal radiation entering the counter would pass 3 mm below the central wire. The design of the side window Geiger tube is shown in Plate II.

The Geiger tube was filled 10 times with an argon-ethylene mixture (9:1) to a pressure of 10 cm of mercury. The average slope of the plateau was 20 percent per 100 volts and the average plateau length was 150 volts. The maximum and minimum slopes were 48 percent and 7.5 percent per 100 volts, respectively. Three fillings using the same ratio of gases by pressure with ethyl alcohol as the quench gas yielded an average slope of 27 percent per 100 volts for the plateau. Chasson (3) found that

EXPLANATION OF PLATE II

Side window x-ray Geiger tube. Made of brass with 0.001 inch aluminum window.

PLATE II


6-No. 12-24 x 1/2" Machine Screws
Spaced on 5 1/16" Diam. Circle
No. 48-4" I.D.
O-RING GROOVE
To Stop-cock
4" Diam.
4 1/4" Diam.
0.003" Tungsten Wire
Stupakoff type GD seal
15/16"
4 3/4"
15/16"
6 1/4" Diam.
6 5/8"
1"
1"
4"
4"

ethyl alcohol usually gives better plateau characteristics than ethylene. It is quite probable that the water vapor content introduced in the alcohol fillings was greater than that in the ethylene fillings as no drying agent was used. The stability of the high voltage that was used on the center wire of the counter, in addition to periodic checks with a standard radioactive source, made the use of fillings with as high as 30 percent plateaus feasible.

For all work in the calibration of the side window Geiger tube, a cobalt x-ray tube, with the tube current stabilized at 5.0 milliamps to within 0.2 percent was used. The double crystal monochromator gave a Co $K_{\alpha 1}$ beam with a divergence of 0.00001 radians of arc. The intensities used were not great enough to be corrected for the 2 millisecond dead time of this counter tube.

Calibration along the length of the window was accomplished by moving the tube perpendicularily to the beam of x-rays by means of a lathe compound. Due to the apparent non-uniformity of the machining of the window opening in the tube, an additional stationary limiting slit was placed in front of and parallel to the counter window. Three curves similar to the one shown in Plate III were obtained by moving the tube carriage 5.1 mm at a time.

In order to determine the effect of the angle of incidence on the measured intensity, the Geiger tube was rotated about an axis at the center of the window. This assured that a variation in the measured intensity upon rotation due to a change in the counter distance from the source was negligible. The limiting slit parallel to the window

EXPLANATION OF PLATE III

The intensity recorded is plotted as a function of the position at which a well collimated and constant x-ray source enters the slit of the side window Geiger tube.

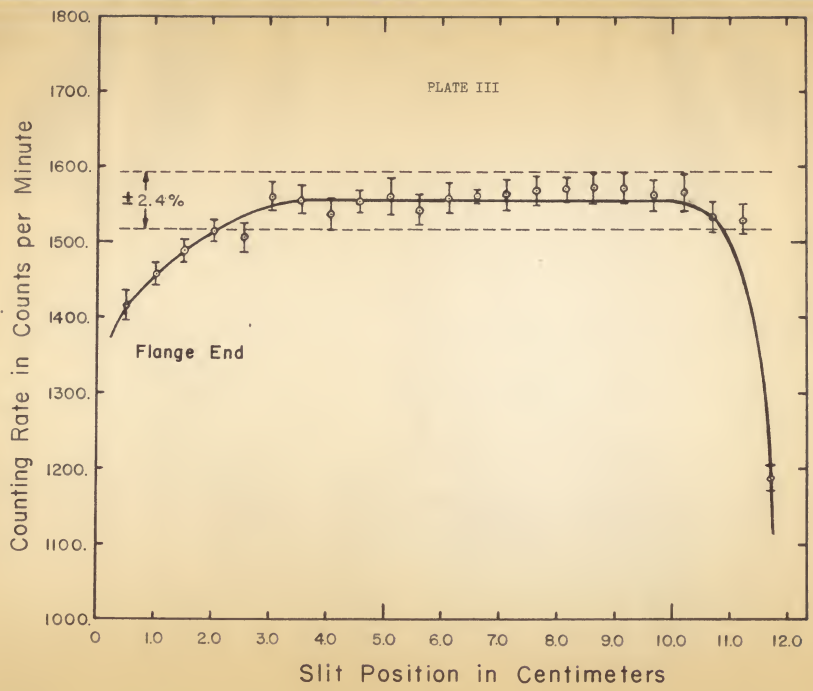


PLATE III
Counting Rate in Counts per Minute
1800.
1700.
1600.
1500.
1400.
1300.
1200.
1100.
1000.
±2.4%
Flange End
0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
9.0
10.0
11.0
12.0
Slit Position in Centimeters

was also employed in these measurements. The intensity was measured as a function of the angle of rotation, in four degree intervals, to 20 degrees on either side of the position where the counter slit was perpendicular to the incoming beam. The variation in counting rate due to counter rotation over this interval was $\pm$ 2 percent.

Although the tube current in the x-ray source was highly stabilized, the high voltage was not. This may have given rise to deviations other than the statistical deviation indicated in Plate III. The results of Plate III indicate that the counting rate is constant to within $\pm$ 2.4 percent, including statistical deviation, over a length of 7.5 cm of the counter. Several trials showed a duplication of the data presented in Plate III and of the results outlined in the above discussion.

In regard to angular calibration, the cylindrical shape of the aluminum window coupled with the need for high precision alignment made it difficult to check data against theoretical predictions. For the Co $K\alpha_1$ radiation, there was an absorption of 47 percent by the aluminum window and of that transmitted through the aluminum, 30 percent was absorbed by the filling gases. It would be expected that in a rotation from normal incidence the intensity measured would be decreased due to the effective increase in thickness of aluminum and increased due to the increase in path length in the tube. The counter-balancing of these two effects probably accounts for the fact that the variation of intensity with angle was as low as indicated above.

From the data collected and presented above, the side window Geiger tube appeared to be quite satisfactory for this problem of finding the

index of refraction by total reflection as the angles involved were much less than the 20 degree limit found in calibration.

The Recording Unit

Pulses from the Geiger tube were fed into a Nuclear Model 1615 Radiation Sentinel which consisted of a high voltage supply for the Geiger tube and a counting rate meter. The counting rate meter was operated on the three percent position. At this position, the meter read the counting rate with a statistical error of three percent. When the meter was used in this manner, the time required for the meter to reach the correct reading was between 30 and 60 seconds.

The output from the counting rate meter was fed through a resistor and potential divider in series. The resistance of this combination, 1350 ohm, is the external resistance for which the meter was designed. A suitable potential was picked off by means of the potential divider and applied to the terminals of a Micromax recording potentiometer. The potentiometer was not calibrated to read counting rate as it was necessary to know only the relative intensities of the reflected beam.

The Nickel Mirror

The nickel mirror was prepared from a solid piece of high purity nickel by Lee (11) in 1949. The mirror had been carefully machined and prepared and only a small amount of polishing on a lapping wheel was necessary to remove the oxide surface that had formed. There were two signi-

ficant reasons for using a solid mirror rather than one made by evaporating the metal onto a piece of plane glass which has been the procedure most commonly used. The first was that the difference in the coefficients of expansion of the nickel and base material would cause strains to be set up in the mirror when the temperature was changed during the investigation. The second was the well known difference in the properties of films of substances from the properties of the substances in bulk (1).

Data Analysis

Alignment was important in determining the intensity of reflected radiation versus glancing angle because of the very manner in which the data were taken. The sample was removed from the sample holder and the table and chamber were placed in a position such that the Co K α_1 beam passed through the windows of the chamber and furnace. With the aid of the Geiger tube and an aligning needle, adjustments were made until it was certain that the beam passed directly over the center of the table. A limiting slit was then placed in a position such that only an unreflected x-ray beam could enter the Geiger tube. Finally, the mirror was placed in the sample holder, the table was rotated and minor adjustments were made until the maximum counting rate shown on the meter was one-half of the value noted without the mirror in place. This procedure assured that the mirror was parallel to and centrally located in the x-ray beam.

As soon as the chamber was evacuated and the sample brought to the

desired temperature, the motion of the cardioid cam and the recording potentiometer were started simultaneously. The curve of reflected intensity versus glancing angle was constructed from the recorder chart with the aid of the calibration curve of glancing angle versus time.

Two curves were obtained from each rotation of the cardioid cam. Shown in Plate IV is an averaged trace of a single curve recorded by the Micromax potentiometer for the nickel mirror at room temperature. The curves of Plate V are those taken from this trace with the aid of the curve of Plate I.

In the case of the trace noted above the intensity was not recorded before the mirror was placed in position. Therefore, in order to determine the zero points* on the curves of Plate V, the maximum intensity of each of these curves was assumed to be 90 percent of the incident intensity (10) and then one-half of the incident intensity intercepted the curve at its zero point. The cutoff position was determined in the usual manner by taking one half of the maximum intensity occurring in that part of the curve where the intensity undergoes its greatest attenuation. For the curves of Plate V, this was one-half of the maximum value recorded. The values of θ_c were then found by taking the difference between the zero position and the cutoff position.

As can be seen from the trace in Plate IV, the mirror began to rotate back through the critical angle before the shape of the cutoff edge

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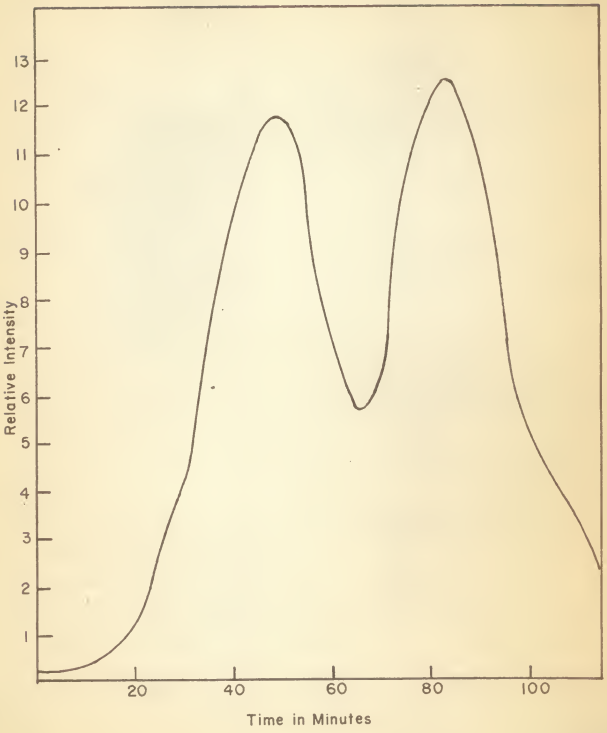
* The term zero point will be used to indicate the angular position, which is arbitrary, at which the mirror is parallel to the incident beam.

EXPLANATION OF PLATE IV

An averaged trace of the curve obtained from the recording Micromax showing the dependance of x-ray intensity on time. The zero of time indicates the simultaneous starting of the Micromax and the rotation of the cardioid cam.

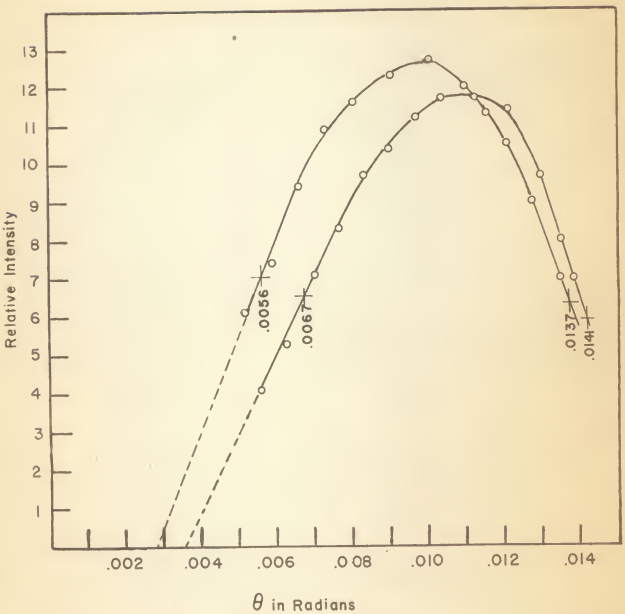
PLATE IV


Relative Intensity
13
12
11
10
9
8
7
6
5
4
3
2
1
20
40
60
80
100
Time in Minutes

EXPLANATION OF PLATE V

A plot of the relative x-ray intensity versus angle of rotation of the nickel mirror. This curve is plotted from the curve of Plate IV with the aid of the curve of Plate I.

PLATE V


Relative Intensity
13
12
11
10
9
8
7
6
5
4
3
2
1
.0056
.0067
.0137
.0141
.002
.004
.006
.008
.010
.012
.014
θ in Radians

was fully determined. From Plate V, it is also noted that the intensity increased beyond the zero points indicated on the curves. This was explained by the fact that the x-ray beam was of finite width. At the zero position, one half of the incident intensity was recorded by the counter tube. Although further rotation of the mirror intercepted more of the beam that by-passed it, total reflection of greater portions of the beam yielded increasingly greater measured intensities until the entire beam fell on the face of the mirror.

EXPERIMENTAL RESULTS

The prime accomplishment of this experiment was the development of the side window Geiger tube. The x-ray intensity was found to be independent of the position at which the radiation entered the window over a length of 7.5 cm. The variation of counting rate was found to be independent of the angle of incidence of the radiation entering the counter to within $\pm$ 2 percent for angles up to 20 degrees. Thus, the need of a mechanism to position a counter in a constant position with respect to the reflected x-ray beam was eliminated.

Circumstances prevented the collection of the volume of data necessary to analyze the problem of the change in the density of free electrons in the vicinity of the Curie temperature. Hence, discussion of the results, errors and conclusions will be confined to the applicability of the apparatus and procedure to the determination of the complex index of refraction.

The only curves obtained were those shown in Plates IV and V. The value of θ_c averaged from these curves was $7.75 \pm 0.35 \times 10^{-3}$ radians. The error here indicated was determined from the deviation of the two values of θ_c from their average value. Errors due to procedure and instrumentation appear below. The value of δ calculated from this value of θ_c was $30.0 \pm 2.6 \times 10^{-6}$. This value of δ is in good agreement with that reported by Lee (11) and Kiessig (9).

The curve of Plate IV was obtained while checking the performance of the Micromax potentiometer. Since extreme care was not given to alignment and positioning, the curve should not be used to judge the performance of the complete apparatus but should serve only to indicate the method of analysis of the data.

Because of the incompleteness of the intensity curve at the critical angle and the errors due to the response of the counting rate meter noted below, no attempt was made to calculate the value of β.

ERRORS AND CONCLUSIONS

The critical angle was obtained from the chart recorded by the Micromax and the calibration curve of the cardioid cam. In the construction of the calibration curve the deflection of the reflected light beam was read on the glass scale to ± 0.05 cm. The distance between the center of the mirror and scale was measured with a steel metric tape to within ± 0.3 cm. As the total distance was found to be 397.7 cm this error was negligible in comparison with the error in read-

ing the deflection. Thus the glancing angle was determined to within $\pm$ 0.06 X 10^{-3} radians.

The time of rotation of the cardioid cam was measured with reference to a small mark scratched on the cam. When the center of the rider passed this mark, the zero of time was recorded. It was estimated that this position could be determined to within $\pm$ 0.5 minutes. The effect of this error is reduced since the calibration curve is very nearly linear, the angular position is only arbitrary, and the critical angle is determined by the difference in the zero and cutoff points. Assuming a value of 7.7 X 10^{-3} radians for θ_c, a few calculations from the calibration curve showed that the maximum error envolved here was $\pm$ 0.05 X 10^{-3} radians.

The greatest error was due to the different rates with which the counting rate meter responded to increasing and decreasing counting rates. From the difference in the slopes of the curves of Plate V, it was determined that the response to a decreasing counting rate was 15 percent faster than the response to an increasing counting rate. This was found to introduce an error of $\pm$ 0.2 X 10^{-3} radians in the determination of the critical angle. Hence, the total error in angle was $\pm$ 0.31 X 10^{-3} radians.

From the data obtained from this experiment the following conclusions were reached:

1. The value of θ_c at room temperature is 7.75 $\pm$ 0.35 X 10^{-3} radians.

2. β cannot be determined from data obtained with the present apparatus because the response of the counting rate meter is not satisfactory.

3. If the rate of oscillation of the mirror were slowed down sufficiently or if a counting rate meter of much faster response were employed, β could be found and θ_c determined to within $\pm$ 0.11 X 10^{-3} radians.

4. The side window Geiger tube appears to be satisfactory for the determination of x-radiation intensities of beams reflected, diffracted or scattered through angles up to 20 degrees.

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Appreciation is expressed to Professor R. D. Dragsdorf, under whose guidance this work was done, for his excellent advice and helpful criticisms throughout the experiment and preparation of this manuscript.

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The apparatus and technique for determining the complex index of refraction of substances at high temperatures were developed. The complex index of refraction was reviewed and its relation to the electron density was emphasized in this work.

A side window Geiger tube was built to alleviate the need for continuously positioning the detector in measuring x-ray intensities deviated through small angles. The Geiger counter had a ± 2.4 percent maximum variation in counting rate, including statistical deviations, along a 7.5 cm length of counter window. Other measurements showed a ± 2 percent variation in counting rate as a function of counter angle for a horizontal rotation of 20 degrees on either side of a position where the counter slit was perpendicular to the incoming beam. The calibration was made using a well-collimated beam of Co $K\alpha_1$ x-rays. The dead time for this Geiger tube was 2 milliseconds.

It was originally intended to investigate the index of refraction of nickel at its Curie temperature because of a change in the free electron density at that temperature which was reported by Cardwell from photoelectric data. Circumstances prevented the completion of this phase of the experiment.

The method and apparatus employed proved successful in determining the real part of the complex index of refraction, $1 - \delta$. With only a minor change in the present apparatus it would be possible to determine the complex part of the index of refraction, $i\beta$. A value of δ at room temperature was found to be $30.0 \pm 2.6 \times 10^{-6}$.